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Integrated torrefaction vs. external torrefaction – A thermodynamic analysis for the case of a thermochemical biorefinery

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Integrated and external torrefaction is analyzed and compared via thermodynamic modeling. In this paper, integrated torrefaction is defined as torrefaction integrated with entrained flow gasification. External torrefaction is defined as the decentralized production of torrefied wood pellets and centralized conversion of the pellets by entrained flow gasification. First, the syngas production of the two methods was compared. Second, the two methods were compared by considering complete biorefineries with either integrated torrefaction or external torrefaction. The first part of the analysis showed that the biomass to syngas efficiency can be increased from 63% to 86% (LHV-dry) when switching from external torrefaction to integrated torrefaction. The second part of the analysis showed that the total energy efficiency (biomass to methanol + net electricity) could be increased from 53% to 63% when switching from external torrefaction to integrated torrefaction. The costs of this increase in energy efficiency are as follows: 1) more difficult transport, storage and handling of the biomass feedstock (wood chips vs. torrefied wood pellets); 2) reduced plant size; 3) no net electricity production; and 4) a more complex plant design.

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1. Introduction

Torrefaction is a low temperature pyrolysis process that can be used as a pretreatment process for biomass. The process can also be referred to as a roasting process and is, e.g., used for roasting coffee beans in the production of coffee. The torrefaction of woody biomass operates typically at 250–300 °C and produces a solid fuel with properties that resemble those of coal. These properties include increased energy density, improved grindability/pulverization, hydrophobic nature, etc. Torrefaction is typically envisioned to be done decentralized, followed by pelletization or briquetting to lower handling, transportation and storage cost. The pretreated biomass can then be sent to a central biomass processing facility [1–3].

Because torrefied biomass has similar properties as coal, it can be gasified using commercial entrained flow coal gasifiers [4,5]. This can enable a relatively quick shift from coal to biomass for syngas production. In Ref. [5], torrefied wood powder is gasified in a 270 kWth oxygen-blown pressurized entrained flow gasifier. The gas produced consist of H_2, CO, CO_2, H_2O and 0.9–1.8 mol% CH_4, together with small amounts of higher hydrocarbons, including tar and soot. A low content of hydrocarbons (~CH_4), and a high content of H_2 and CO, is preferable when using the syngas for chemical synthesis. It is concluded in Ref. [5] that torrefaction reduces the CH_4 content in the syngas, and it is shown that increasing the torrefaction temperature from 300 °C to 340 °C lowers the CH4 content, but at the cost of a lower cold gas efficiency [5]. The disadvantage of entrained flow gasification of torrefied biomass is the relatively low cold gas efficiency achieved due to the high temperature in the gasifier. When also considering the loss of heating value in the torrefaction step the "overall cold gas efficiency" becomes even lower. A method to increase the cold gas efficiency of entrained flow gasification of torrefied biomass has been presented by Mark Prins et al. [4]. The method is about integrating the torrefaction step with the entrained flow gasification. This means that the torrefaction process is done centralized at the gasifier and not decentralized, resulting in higher transportation, handling and storage cost of the untreated biomass compared to torrefied biomass pellets [6].

This paper will compare centralized torrefaction with the more conventional decentralized torrefaction, or when the perspective of a centralized plant is taken; compare external torrefaction to

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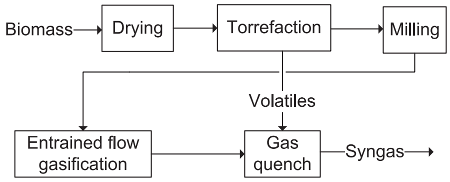

Fig. 1. Simplified flow sheet showing the idea behind integrated torrefaction as presented in Ref. [4]. Note: for case c and d presented in the text: biomass = wood chips (willow).

integrated torrefaction. This paper will add to the knowledge that is available in the open literature about integrated torrefaction, by building on top of the analysis performed by Prins et al. in Ref. [4]. This paper will examine the integration between torrefaction and gasification more closely – mainly concerning the increase in the overall cold gas efficiency. Some of the questions answered include: what causes the increase in the overall cold gas efficiency? How do torrefaction conditions influence the increase in overall cold gas efficiency? How does the overall cold gas efficiency compare with external torrefaction or no torrefaction? These questions are answered by thermodynamic modeling of the biomass to syngas conversion (torrefaction + gasification). The integrated torrefaction process will however benefit from heat integration with the downstream syngas conversion processes; such as the conversion of syngas to fuel product, or syngas to electricity. Because of this, the paper will also show the impact of using either external torrefaction or integrated torrefaction in a thermo-chemical biorefinery via thermodynamic modeling and analysis. The impact is assessed by comparing the fuel energy efficiencies as well as the total energy efficiencies along with an assessment of the complexity of the plant designs. Many studies have been published on modeling and analysis of thermo-chemical biorefineries, such as [7–11], but none have used integrated torrefaction.

2. Integrating torrefaction with entrained flow gasification

The concept of integrated torrefaction is presented in Fig. 1. It shows how the volatile gasses produced in the torrefaction process can be converted to syngas when using entrained flow gasification [4].

In decentralized torrefaction, the volatile gasses are typically combusted to produce heat for drying and torrefaction [2]. In a centralized plant, the use of medium and low temperature waste heat for drying and torrefaction is more optimal, and the volatile gases can then be used to chemically quench the high temperature gas from the gasifier and thereby increase the production of syngas.[1] The method was proposed by Mark Prins et al. in Ref. [4]. Mark Prins et al. write that "due to the high temperature, the thermally unstable volatiles from the torrefaction step will decompose into carbon monoxide and hydrogen" [4]. This claim is the core of the integrated torrefaction process and is therefore also a prerequisite for this paper. It should be noted that the claim still needs to be verified by experiments. If future experiments show that the volatiles are not decomposed into hydrogen and carbon monoxide to a very significant extent, the method would probably not be attractive, especially if significant amounts of tar compounds in the volatiles from torrefaction "survive" the high temperature quench. If this shows to be the case, a dedicated downstream tar reforming/cleaning process, similar to what is used when gasification is done in fluidized beds, would be required. A dedicated downstream tar reforming/cleaning process would normally not be needed after an entrained flow gasifier [5,12].[2]

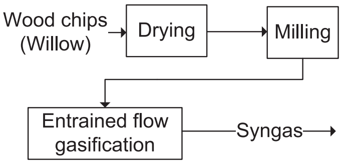

Fig. 2. Simplified flow sheet of syngas production from willow without torrefaction. Note: Untreated willow is assumed to be able to be gasified in an entrained flow gasifier. Moreover, the same gasification conditions as those used for torrefied wood are assumed applicable.

To evaluate the integrated torrefaction process, a parameter called the "overall cold gas efficiency" is defined as follows:

$$\text{overall cold gas efficiency} = \frac{\dot{m}_{\text{syngas}} \cdot \text{LHV}_{\text{syngas}}}{\dot{m}_{\text{biomass,dry}} \cdot \text{LHV}_{\text{biomass,dry}}} \tag{1}$$

This parameter is the total energy efficiency for the integrated system, which consists of torrefaction and gasification (and gas quench).

In Section 2.2, the values for the overall cold gas efficiency are presented for four cases. The cases are a) without torrefaction (Fig. 2), b) external torrefaction (Fig. 3), c) integrated torrefaction at 250 °C (Fig. 1) and d) integrated torrefaction at 300 °C (Fig. 1). The modeling approach used to generate the results for the four cases is presented in Section 2.1.

2.1. Modeling of torrefaction and gasification

Torrefaction is modeled in a certain way for the case of external torrefaction and in another way for the cases of integrated torrefaction. For external torrefaction, simply using the energy efficiency of torrefaction is adequate (Eq. (2)), but for integrated torrefaction, torrefaction is modeled in more detail based on two torrefaction experiments presented in Ref. [4].

$$\text{energy efficiency of torrefaction} = \frac{\dot{m}_{\text{torr\_wood}} \cdot \text{LHV}_{\text{torr\_wood}}}{\dot{m}_{\text{biomass,dry}} \cdot \text{LHV}_{\text{biomass,dry}}} \tag{2}$$

The two experiments are summarized in Fig. 4. The two experiments estimate the reaction heat for torrefaction to be lower than 1% of the LHV of the wood input, but the estimation is associated with an uncertainty of 2.2%–2.5% of the LHV of the wood input. The two experiments also show that only 1% of the heating value in the wood input is lost in the torrefaction process (converted to sensible heat).

For external torrefaction, the energy efficiency of torrefaction is set to 80% (dry LHV) irrespective of the torrefaction temperature. This setting agrees with a review on torrefaction [1] and with data on the production of torrefied wood pellets [14,15].[3] An energy efficiency of torrefaction of 80% also matches the torrefaction

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[1] When pressurized entrained flow gasification is used, the volatiles must be pressurized before the gas quench (Fig. 1). This pressurization can either be done by 1) pressurizing the volatiles after torrefaction, 2) pressurized torrefaction, 3) a combination of options 1 and 2.

[2] This is true for coal gasification [5,13], and experiments performed in Refs. [5,12] with wood and torrefied wood indicate the same.

[3] The torrefaction energy efficiency is close to 80% for external torrefaction irrespective of the torrefaction temperature because of the heat requirement for drying. The heat requirement for drying is independent of the torrefaction temperature and much higher than the heat requirement for torrefaction. In many references, torrefaction efficiencies higher than 80% are stated, e.g., Refs. [14,15]. The efficiencies in these references are based on LHV on wet basis instead of dry basis. This difference can clearly be seen in table 2.2 in Ref. [15] and figures 4.4 and 4.5 in Ref. [14]. One of the consequences of basing the energy efficiency on LHV wet basis is that the process of drying wood will produce heating value, even though chemical reactions do not take place.

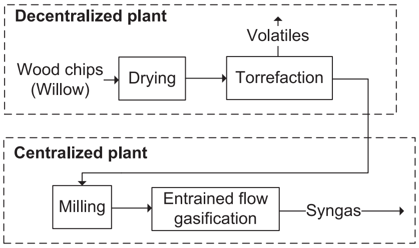

Fig. 3. Simplified flow sheet of syngas production from willow with external/decentralized torrefaction. Note: The torrefied product would be densified by pelletization or briquetting before transportation. This process is not considered in this simplified approach.

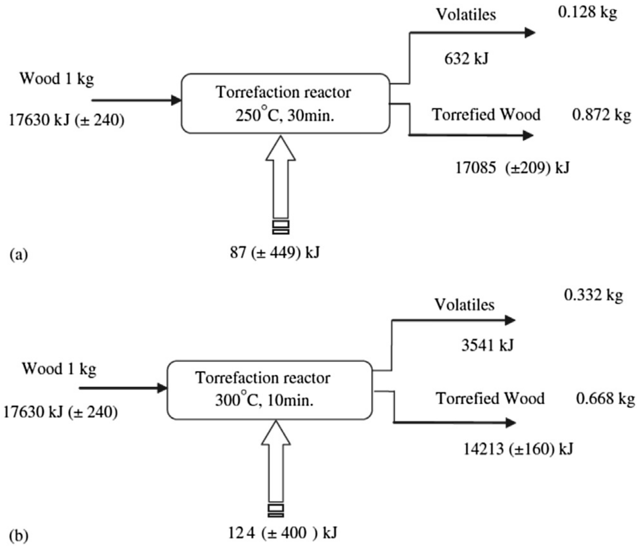

Fig. 4. Overall mass and energy balances for torrefaction of dry willow at temperature and reaction time of (a) 250 °C and 30 min (b) 300 °C and 10 min. Figure from Ref. [4]. The energy content given for volatiles and torrefied wood is a sum of chemical energy and sensible heat. Note: In Ref. [4], the content of chemical energy and sensible heat is also stated separately.

Table 1
Dry composition (in wt%) of solid fuels used in the analysis.

	Wood chips (willow)	Torrefied wood (250 °C, 30 min)	Torrefied wood (300 °C, 10 min)	Torrefied wood pellets[a]
Carbon	47.2	51.3	55.8	50.8
Hydrogen	6.1	5.9	5.6	5.8
Oxygen	45.1	40.9	36.2	41.4
Nitrogen	0.3	0.4	0.5	0.3
Sulfur	0.02[b]	0.02[c]	0.03[c]	0.06
Ash	1.3	1.5	1.9	1.7
LHV (dry basis) [MJ/kg]	17.6	19.4	21.0	20.6
Moisture content [wt%]	5.0[d]	0	0	3.0
c_p [kJ/(kg K)]	1.35[e]	1.28[f]	1.18[f]	1.35[e]
Reference	[4]	[4]	[4]	[2,16]

[a] The torrefied wood pellets are assumed to be the fuel produced in the case of “external torrefaction”.
[b] Sulfur content from [28].
[c] Calculated based on[b].
[d] Assumed value after drying (before torrefaction).
[e] Assumed value.
[f] Calculated based on[e].

efficiency from Ref. [4] at 300 °C, and [14] shows that the energy content of the volatiles is sufficient to meet the heat demand for drying and torrefaction when torrefaction proceeds at 300 °C. It should however be noted that the energy efficiency of torrefaction at a lower temperature (e.g., 250 °C) can be higher than 80% if the energy demand for drying (and torrefaction) is met by using external waste heat instead of burning some of the biomass.

The entrained flow gasification process is modeled by assuming chemical equilibrium at the exit of the gasifier. Due to the high temperature of 1300 °C at the exit of the gasifier, this assumption is good [4,13,17].[4] The process parameters set for the gasification process are shown in Table 3 under “gasifier”.

The chemical quench that occurs when the hot gas from the gasifier is mixed with the volatiles from torrefaction is modeled by assuming that the volatiles are completely converted to H_2, CO, H_2O and CO_2, as suggested by Ref. [4], and that these four gas components are at chemical equilibrium at the exit temperature and pressure of the quench. These assumptions are most accurate at high temperature, which corresponds to a low mass flow of volatiles (low torrefaction temperature). Gas compositions before and after the chemical quench can be seen in Table 4 (integrated torrefaction), and in Table 5 the gas composition of the gas from the gasifier in the case of external torrefaction can be seen.

2.2. Results and discussion

The results show that the highest overall cold gas efficiency is 86% for integrated torrefaction at a torrefaction temperature of 300 °C. The lowest overall cold gas efficiency is 63% for external torrefaction (Fig. 5). This difference is mainly due to the utilization of the volatiles from torrefaction. In integrated torrefaction, the volatiles are converted to syngas, whereas the volatiles are combusted and the heat used for drying and torrefaction in external torrefaction. Because the volatiles contain 19% of the heating value of the input wood when torrefaction is performed at 300 °C, the impact of combusting the volatiles instead of producing syngas is significant.

The case without torrefaction achieves an overall cold gas efficiency of 76%. Therefore, integrating torrefaction can increase the overall cold gas efficiency by up to 10%-points (to 86%). The 10%-point increase can be split into four categories, as shown in Table 2. The most important categories for integrated torrefaction at 300 °C are “volatiles avoiding gasifier” (4.6%-points) and “increase in cold gas efficiency of gasifier” (4.2%-points). For integrated torrefaction at 250 °C, the “increase in cold gas efficiency of gasifier” (3.9%-points) accounts for almost all of the increase in overall cold gas efficiency (increase of 4.5%-points). The chemical quench that occurs when the volatiles are mixed with the hot gas from the gasifier also provides an important contribution to the overall cold gas efficiency for the case of integrated torrefaction at 300 °C (2.2%-points), whereas the contribution for the case of integrated torrefaction at 250 °C is much lower (0.5%-points). This difference is expected, as the volatiles contain almost six times more energy when torrefaction occurs at 300 °C compared to 250 °C (Fig. 5). The

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[4] The methane content may be higher than predicted by chemical equilibrium as indicated in Ref. [5]. The experiments in Ref. [5] are done on a small gasifier (270 kWth). It is expected that increasing the gasifier size to commercial scale, which could be 1000 times bigger (270 MWth), would lower the methane content. When using coal instead of biomass, the methane content is lower than predicted by chemical equilibrium [13].

Table 2
The effect on the overall cold gas efficiency of adding torrefaction.

	Without torrefaction	External torrefaction	Integrated torrefaction, 250 °C	Integrated torrefaction, 300 °C
Cold gas efficiency of gasifier [%]	76.1	79.7	80.2	81.4
Overall cold gas efficiency [%]	76.1	63.4	80.7	86.3
Increase in overall cold gas efficiency compared with “without torrefaction” [%-points]	–	−12.8	4.5	10.2
The total increase given in the row above is split into increments in the following rows [%-points]:				
1. Increase in cold gas efficiency of gasifier	–	2.8	3.9	4.2
1.1 Decrease in moisture content of fuel[a]	–	0.6	1.6	1.4
1.2 Temperature change of fuel[a,b]	–	−0.4	1.1	1.2
1.3 Change in fuel composition (LHV, c_p, ultimate analysis)[a]	–	2.6	1.2	1.7
2. Volatiles avoiding gasifier	–	–	0.8	4.6
3. Chemical quench	–	–	0.5	2.2
4. Loss of LHV in torrefaction	–	−15.6	−0.7	−0.9

[a] The splitting between 1.1 and 1.3 is done by doing parameter variation in the thermodynamic model.
[b] A temperature of 115 °C is assumed in “without torrefaction”, 60 °C in “external torrefaction”, 250 °C or 300 °C in integrated torrefaction.

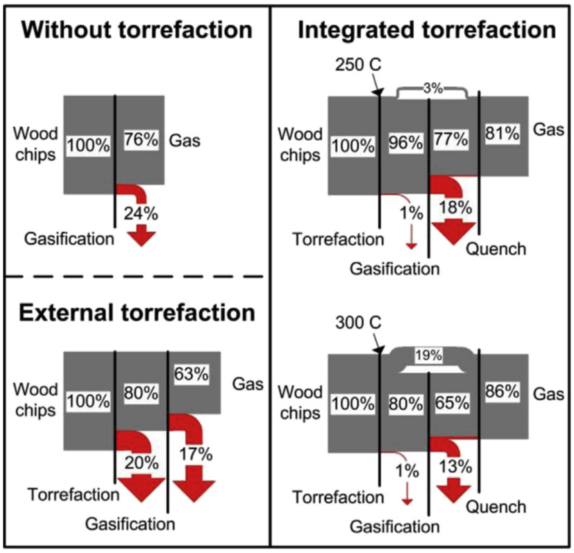

Fig. 5. Sankey diagram of the chemical energy flows and conversion heat losses for the four different cases. The chemical energy flows are based on LHV (dry basis).

results achieved for the cases of integrated torrefaction match well with the data from Ref. [4].[5]

3. Design of the biorefineries

The analysis presented in the previous section of the paper for the cases of integrated torrefaction assumed that the heat demand for drying and torrefaction was met by available waste heat in the downstream processing. This part of the paper investigates the correctness of this assumption by considering the case of a thermochemical biorefinery. The two cases, external torrefaction and integrated torrefaction at 300 °C, are therefore expanded to show the effect on plant design and energy efficiencies of using either external or integrated torrefaction in a biorefinery.

The simplified flow sheets of the two investigated biorefineries are shown in Fig. 6 and Fig. 7. The biorefineries are modeled in Aspen Plus and an in-house modeling tool called DNA [18].[6] The design process parameters used in the modeling are presented in Table 3. Important parts of the plant design are presented below together with the modeling approach.[7] The biorefinery based on external torrefaction is to a great extent based on a biorefinery producing *dimethyl ether* (DME) presented in Ref. [10].

3.1. *Size of the biorefineries*

The size of the biorefinery based on torrefied wood pellets is set to 2312 MWth input as in Ref. [10]. The size of the biorefinery based on wood chips is reduced to 500 MWth input because wood chips are more bulky and have a lower energy density than torrefied wood pellets. Therefore, they are more expensive to transport over very long distances. For comparison, some kraft pulp and paper mills are bigger than 500 MWth [19]. Because this study does not include an economic analysis, the exact sizes of the plants do not impact the results and conclusions of the paper.

3.2. *Pretreatment & feeding*

The torrefied wood is assumed to be pretreated and fed with existing commercial coal technology [2,3]. The torrefied wood pellets are milled to powder, and the powder is pressurized with lock hoppers and fed to the gasifier with pneumatic feeders, both of which use CO_2 from the carbon capture process downstream. The wood chips are fist dried in a steam dryer, then pressurized with lock hoppers, torrefied at 300 °C and milled to powder, which is fed to the gasifier with pneumatic feeders.[8]

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[5] From Table 3 in Ref. [4], an exergy efficiency from wood to product gas can be calculated to be 15,178/20023 = 75.8% for case IIIa and 16,050/20,023 = 80.2% for case IIIb (entrained flow gasification at 1200 °C integrated with torrefaction at 250 °C (IIIa) or 300 °C (IIIb)). Fig. 6 in Ref. [4] shows that the physical exergy content of the gas is negligible because it is calculated after the gas cooler/HRSG. If a wood to gas chemical exergy efficiency is calculated for the present cases, the results are 73.6% and 78.5% for torrefaction at 250 °C and 300 °C, respectively. If the model is changed so that entrained flow gasification is performed at 1200 °C instead of 1300 °C, the results become 75.0% and 79.5% for torrefaction at 250 °C and 300 °C, respectively.

[6] The gasification part is modeled in DNA and the rest of the biorefinery in Aspen Plus. The property methods used in Aspen Plus are: PR-BM (Peng-Robinson with Boston-Mathias alpha function) for the syngas processing, SR-POLAR (Schwarzentruber and Renon) for the methanol reactor loop and the distillation, and STEAMNBS for the integrated steam plant.

[7] The modeling approach for the torrefaction part is described in Section 2.1 above. It is only the torrefaction part that is directly based on experimental data, the rest of the plants consists of conventional components that can be modeled by using the process design parameters given in Table 3.

[8] Could also be torrefied at lower pressure and then pressurized further after milling, but this approach would also mean that the volatiles from torrefaction would need to be pressurized at high temperature to the gasification pressure.

Table 3
Main process design parameters used in the modeling.

Feedstock	Torrefied wood pellets or wood chips – compositions are given in Table 1
Steam dryer	Water content after dryer = 5wt%
Pretreatment	Power consumption for milling = 0.29% of the thermal input (LHV)[a]
Pressurizing & Feeding	Pressurizing: CO2 for pressurizing is assumed to be reused (no net CO_2 consumption). Feeding: CO_2/biomass mass-ratio = 22%
Torrefaction	Outlet temperature = 300 °C. Heat demand = 2% of biomass energy input (LHV)[b].
Gasifier	$P_{exit} = 45$ bar [20]. ΔP = 1.2 bar. Temp. before quench = 1300 °C[c]. Carbon conversion = 100% [10]. Heat loss: 2.7% of the thermal input is lost to surroundings and 1% of the thermal input is used to generate steam [10]. Chemical equilibrium (Gibbs minimization) at the exit temperature and pressure. c_p for ash = 1 kJ/(kg K)
Air separation unit	O_2 purity = 99.6 mol%. Electricity consumption = 1.0 MWe/(kg-O_2/s) [27]
Water gas shift (WGS) reactor	Pressure drop = 2 bar. Chemical equilibrium of the WGS reaction at the reactor exit temperature (383–440 °C).
AGR (Rectisol)	Power consumption = 0.11 MW/($kg_{gas\text{-}input}$/s). Heat consumption = 0.074 MW/($kg_{gas\text{-}input}$/s) [10]
Methanol synthesis	Boiling water reactor (isothermal). Reactor inlet: $H_2/CO = 2$, $CO_2 = 3$ mol%. Reactor outlet: $T = 260$ °C, $P = 80.7$ bar $\Delta P_{reactor} = 4.3$ bar. Approach to equilibrium of 15 °C for both the methanol reaction (Eq. (3)) and the WGS reaction [11].
Distillation	$P = 8$ bar (topping column), 1.0 bar (methanol column). CO_2 content in liquid after topping column = 1 ppm. Water content in product methanol = 0.1 mol%. Methanol content in bottoms product = 1–10 mol % Number of stages in distillation columns: 10 (topping column), 35 (methanol column). Feed stage position optimized.
Heat exchangers	$\Delta T_{min} = 30$ °C (gas–gas) or 10 °C (gas – liq. and liq. – liq.) or 5 °C (lig. – evap./cond.-liq.).
Steam cycle	Steam temperature before turbine = 600 °C. Maximum steam pressure = 180 bar. Minimum steam quality after turbine = 0.85. Condensation pressure = 0.042 bar. Isentropic efficiency of turbine = 0.9
Gas turbine	Air compressor: pressure ratio = 19.5[d], $\eta_{polytropic} = 87\%$[d]. Turbine: TIT = 1370 °C[d], $\eta_{isentropic} = 89.8\%$[d] $\eta_{mechanical} = 98.7\%$[d] $\eta_{electrical} = 98.6\%$[d]
Compressors	$\eta_{polytropic} = 85\%$ (3 stage O_2 compression from 1 to 46 bar and 5 stage CO_2 compression from 1 to 46 bar), 80% (syngas compressors)[d] $\eta_{mechanical} = 94\%$[d] $\eta_{electrical} = 100\%$

[a] [23]. In Ref. [2], the power consumption for milling torrefied biomass and bituminous coal were experimentally determined to be the same (1% of the thermal input). The size of the mill used in the experiments (heavy-duty cutting mill, 1.5 kWe) was assumed to be the reason for the higher value (1% vs. 0.29%).

[b] The heat demand for torrefaction at 300 °C was lower than 1% of the biomass energy input (LHV) according to [4] – see also Fig. 4. It is conservatively set to 2% in the modeling (also accounts for heat losses).

[c] In Ref. [16], 1300 °C was used for the entrained flow gasification of torrefied biomass. The addition of silica or clay to the biomass is likely necessary at this relatively low temperature to make the gasifier slag [16], but these compounds are not added in the modeling. The enthalpy of fusion of the ash is also not included in the modelling.

[d] [23]. Note for gas turbine: The gas turbine is a natural gas fired gas turbine (GE 7FB) that is fitted to use syngas. In Ref. [23], the simulations of the gas turbine operating on syngas show that the $m_{air\ compressor}/m_{turbine}$ ratio can be 0.91 – in this paper, the ratio is 0.93–0.94. This high ratio is a result of the composition of the unconverted syngas (contains 56–61 mol% H_2). Typically, the TIT would be de-rated by 20–30 °C when operating on syngas (compared to natural gas) or up to 50 °C when operating on hydrogen. However, (as suggested in Ref. [23]), the historic increase in TIT is assumed to continue. Therefore, the TIT of 1370 °C has not been de-rated.

3.3. *Gasification*

A commercial, dry-fed, slagging[9] entrained flow coal gasifier is used to gasify the torrefied wood powder. The gasifier is oxygen blown and pressurized to 45 bar [20]. A cryogenic air separation unit provides the oxygen supply. A gas quench using 200 °C recycled syngas downstream of the dry solids removal lowers the temperature of the syngas from 1300 °C to 900 °C for the biorefinery using external torrefaction. For the biorefinery using integrated torrefaction, the temperature is lowered by quenching the syngas at 1300 °C with the volatiles from the torrefaction process upstream. The composition of the syngas after the gasifier and after the chemical quench with the volatiles is calculated by assuming chemical equilibrium at the exit temperature and pressure (compositions given in Tables 4 and 5).

3.4. *Gas cooling and water gas shift*

The syngas is further cooled to 200 °C by generating superheated steam[10]. A sulfur tolerant[11] WGS (water gas shift) reactor adjusts the H_2/CO ratio to two to optimize methanol synthesis according to Eq. (3). After the WGS reactor, the gas is cooled prior to the acid gas removal step.

3.5. *Gas cleaning*

The gas cleaning of biomass syngas for methanol synthesis includes cyclones and filters for particle removal placed just after the high temperature syngas cooler, an AGR (acid gas removal) step and guard beds[12] placed just before the synthesis reactor [23,24]. The AGR step is performed with a chilled methanol process similar to the Rectisol process [10], and it removes sulfur components (H_2S and COS[13]), CO_2 and other species, such as NH_3 and HCl. CO_2 is vented to the atmosphere and the sulfur components are sent to the gas turbine. The H_2S + COS content in the syngas after AGR is approximately 0.1 ppm [25], and the CO_2 content is 1.2–1.5 mol%.[14]

The energy input for the AGR process is primarily electricity to power a cooling plant, but electricity is also used to run pumps that pressurize the methanol solvent. A heat input is used to cover the reboiler duty of the water removal column and the sulfur removal column.

3.6. *Synthesis of methanol*

The syngas is compressed to 85 bar prior to entering the synthesis reactor. The reactor is modeled as an isothermal boiling water reactor that operates at 260 °C, where the methanol reaction (Eq. (3)) and the water gas shift reaction are assumed to reach an approach to equilibrium of 15 °C [26]. The reactor product gas is cooled to 40 °C to condense 98–99% of the produced methanol. A

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[9] Because of the low ash content in biomass, a slag recycle is needed to ensure that the gasifier slags [16]. Moreover, see note c below Table 3.

[10] Steam is superheated to 600 °C in the gas cooling. Reference [20] states that only a “mild superheat” can be used in the gas cooling, but in Ref. [21] steam at 125 bar is superheated to 566 °C.

[11] E.g. Haldor Topsoe produces such catalysts [22].

[12] ZnO and active carbon filters.

[13] Sulfur is only modeled as H_2S.

[14] The CO_2 concentration before the methanol reactor is set to 3 mol% to ensure catalyst activity in the methanol reactor [13]. The remaining CO_2 will be supplied by the recycled unconverted syngas.

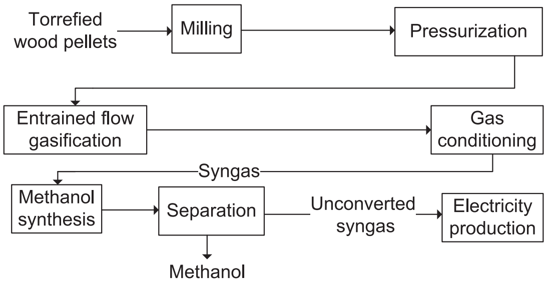

Fig. 6. Simplified flow sheet of the methanol plant based on torrefied wood pellets (external torrefaction).

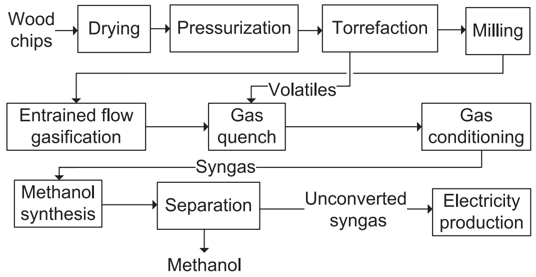

Fig. 7. Simplified flow sheet of the methanol plant based on wood chips (integrated torrefaction).

gas–liquid separator separates the liquid from the unconverted syngas, 97% (external torrefaction) or 88% (integrated torrefaction) of the unconverted syngas is recycled to the synthesis reactor. The remaining gas is sent to the gas turbine.

$$2H_2 + CO \leftrightarrow CH_3OH \tag{3}$$

3.7. Distillation

The liquid stream from the gas–liquid separator is distilled by fractional distillation in two columns. The first column is a topping column that separates the absorbed gasses from the liquid. The gas from the topping column, which consists mainly of CO_2, is compressed and recycled back to the AGR. The second column separates water from methanol. The methanol liquid product achieves a purity of 99.9 mol%. The feed to the distillation columns is preheated (partly evaporated) in most cases. This preheating lowers the reboiler duty and enables the utilization of lower temperature waste heat.

3.8. Heat integration and electricity production

The heat integration within the biorefinery based on integrated torrefaction is more comprehensive and complex compared to the biorefinery based on external torrefaction. This complexity is due to the heat demand for drying and torrefaction. Therefore, the waste heat available for the integrated steam cycle is significantly reduced. The electricity production from the integrated steam cycle is supplemented by a gas turbine that operates on unconverted syngas. In the biorefinery based on integrated torrefaction, the gas turbine produces almost the same amount of electricity as the steam cycle to meet the on-site electricity consumption. In the biorefinery based on external torrefaction, the gas turbine produces 14 times less electricity than the steam cycle.

4. Results

The results from the thermodynamic modeling of the two biorefineries are presented below. The flow sheets of the two plants are given in Fig. 8 and Fig. 9, accompanied by Tables 4 and 5 with the compositions of selected mass flows. Table 6 compiles the main results. The results show higher energy efficiency in the biorefinery that uses integrated torrefaction compared to the biorefinery that uses external torrefaction. This difference was expected based on the results presented in Section 2, which showed an increase in the biomass to syngas efficiency (overall cold gas efficiency) of 36% (from 63% to 86%) when converting from external torrefaction to integrated torrefaction. The calculated biomass to methanol + net electricity efficiencies is 53% for the biorefinery that uses external torrefaction, and 63% for the biorefinery that uses integrated torrefaction. This constitutes an increase of 20% when converting from external torrefaction to integrated torrefaction. This shows that the increase of 36% in energy efficiency for the syngas production (overall cold gas efficiency) is reduced to 20% when considering the total energy efficiency of the biorefineries. This reduction is mainly due to the heat requirement for drying and torrefaction, which lowers the on-site electricity production from the integrated steam cycle from 11% (external torrefaction) to 3% (integrated torrefaction) of the biomass/torrefied biomass energy input. Therefore, the net electricity production is 0% for the biorefinery based on integrated torrefaction, whereas it is 4% of the biomass energy input for the biorefinery based on external torrefaction. To cover the on-site electricity consumption in the biorefinery based on integrated torrefaction, the recycle ratio in the synthesis loop had to be reduced from 97% (external torrefaction) to 88% (integrated torrefaction) to increase the gas turbine electricity production.

Fig. 10 shows the chemical energy flows from wood chips to methanol for the two biorefineries. The figure clearly shows that the conversion heat losses for the syngas production part are much smaller for integrated torrefaction compared to external torrefaction, which was also shown in Section 2 of this paper. The figure also shows that the unconverted syngas flow to the gas turbine is increased from 2% (external torrefaction) to 7% (integrated torrefaction) of the biomass input. This increase aimed to meet the on-site electricity consumption.

Fig. 11 shows the carbon flows in the two biorefineries. The carbon flows to some extent resemble the energy flows from Fig. 10.[15] The majority of the carbon from the biomass is converted from CO to CO_2 in the water gas shift (WGS) and then removed in the acid gas removal (AGR). Some of this CO_2 is used to feed the biomass to the pressurized gasifier, and the rest is vented to the atmosphere. This pure CO_2 stream could be pressurized and stored underground as done in Ref. [10]. This process would increase the on-site electricity consumption, which would result in lower methanol production for the biorefinery based on integrated torrefaction because more syngas would be needed for the gas turbine to meet the added electricity consumption.

5. Discussion

The main results from the analysis of the two biorefineries can be summarized in the following: Switching from external torrefaction to integrated torrefaction will increase energy efficiency, but at the cost of a more complex plant design, reduced plant size,

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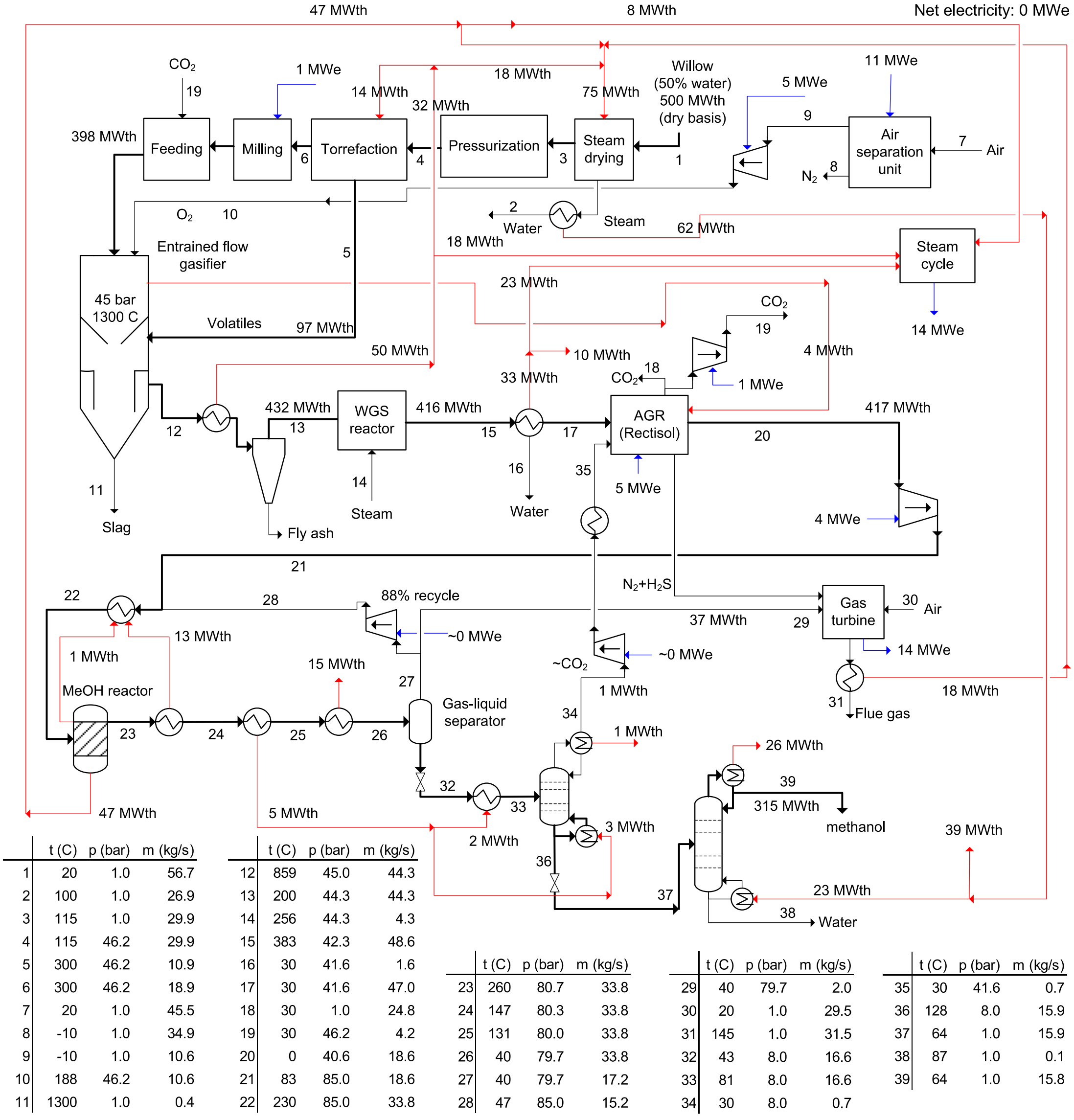
[15] The loss of carbon in the torrefaction for the case of external torrefaction is an example: the loss of carbon matches the loss in chemical energy almost completely (21% vs. 20%).



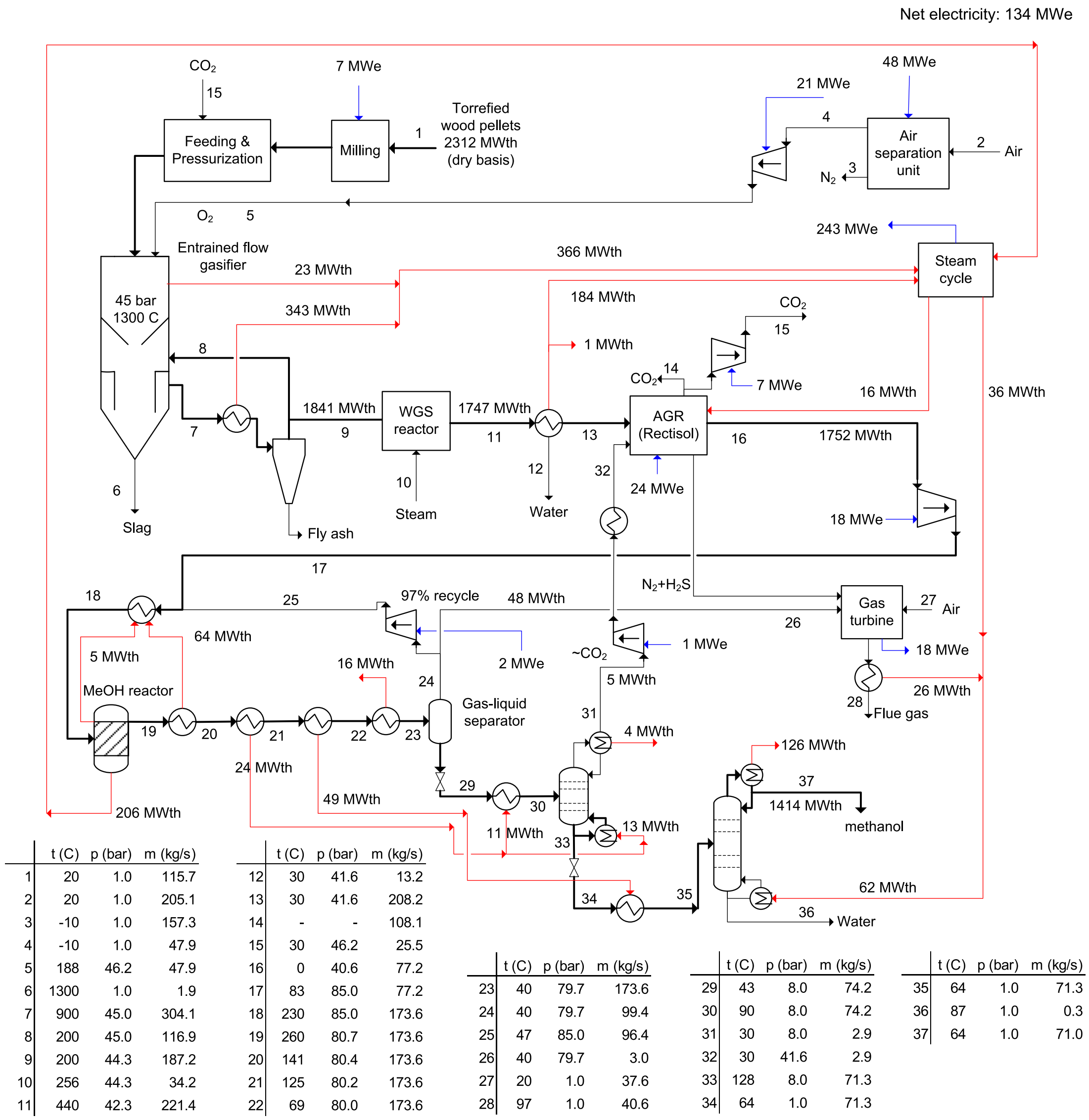
	t (C)	p (bar)	m (kg/s)
1	20	1.0	56.7
2	100	1.0	26.9
3	115	1.0	29.9
4	115	46.2	29.9
5	300	46.2	10.9
6	300	46.2	18.9
7	20	1.0	45.5
8	-10	1.0	34.9
9	-10	1.0	10.6
10	188	46.2	10.6
11	1300	1.0	0.4
12	859	45.0	44.3
13	200	44.3	44.3
14	256	44.3	4.3
15	383	42.3	48.6
16	30	41.6	1.6
17	30	41.6	47.0
18	30	1.0	24.8
19	30	46.2	4.2
20	0	40.6	18.6
21	83	85.0	18.6
22	230	85.0	33.8
23	260	80.7	33.8
24	147	80.3	33.8
25	131	80.0	33.8
26	40	79.7	33.8
27	40	79.7	17.2
28	47	85.0	15.2
29	40	79.7	2.0
30	20	1.0	29.5
31	145	1.0	31.5
32	43	8.0	16.6
33	81	8.0	16.6
34	30	8.0	0.7
35	30	41.6	0.7
36	128	8.0	15.9
37	64	1.0	15.9
38	87	1.0	0.1
39	64	1.0	15.8

Fig. 8. Detailed flow sheet of the methanol plant based on wood chips (integrated torrefaction). The heat integration as well as power consumption and production are shown in the flow sheet.

Table 4
Stream compositions for the methanol plant shown in Fig. 8 (integrated torrefaction).

	Gasifier (before quench)	Gasifier exit	WGS outlet	AGR outlet	Reactor inlet	Reactor outlet	Purge gas	To distillation	Recycle CO_2
Stream number	–	12	15	20	22	23	29	32[a]	34
Mass flow (kg/s)	33.4	44.3	48.6	18.6	33.8	33.8	2.0	16.6	0.7
Mole flow (kmole/s)	1.50	2.15	2.39	1.66	2.81	1.82	0.15	0.51	0.02
Mole frac (%)									
H_2	23.4	32.5	45.4	65.6	63.5	43.5	60.5	0.29	8.6
CO	55.7	43.0	22.5	32.6	31.8	22.0	30.6	0.26	7.8
CO_2	9.1	13.1	28.0	1.5	3.0	4.5	5.2	2.7	79.1
H_2O	11.2	11.2	3.9	–	0.00	0.17	0.00	0.59	0.00
CH_4	0.02	0.02	0.02	0.03	0.11	0.17	0.24	0.01	0.32
H_2S	0.01	0.01	0.01	–	–	–	–	–	–
N_2	0.20	0.14	0.13	0.19	0.92	1.4	2.0	0.02	0.51
Ar	0.09	0.06	0.06	0.09	0.41	0.63	0.87	0.03	0.76
CH_3OH	–	–	–	0.03	0.29	27.6	0.67	96.1	2.9

[a] Liquid.



	t (C)	p (bar)	m (kg/s)
1	20	1.0	115.7
2	20	1.0	205.1
3	-10	1.0	157.3
4	-10	1.0	47.9
5	188	46.2	47.9
6	1300	1.0	1.9
7	900	45.0	304.1
8	200	45.0	116.9
9	200	44.3	187.2
10	256	44.3	34.2
11	440	42.3	221.4
12	30	41.6	13.2
13	30	41.6	208.2
14	-	-	108.1
15	30	46.2	25.5
16	0	40.6	77.2
17	83	85.0	77.2
18	230	85.0	173.6
19	260	80.7	173.6
20	141	80.4	173.6
21	125	80.2	173.6
22	69	80.0	173.6
23	40	79.7	173.6
24	40	79.7	99.4
25	47	85.0	96.4
26	40	79.7	3.0
27	20	1.0	37.6
28	97	1.0	40.6
29	43	8.0	74.2
30	90	8.0	74.2
31	30	8.0	2.9
32	30	41.6	2.9
33	128	8.0	71.3
34	64	1.0	71.3
35	64	1.0	71.3
36	87	1.0	0.3
37	64	1.0	71.0

Fig. 9. Detailed flow sheet of the methanol plant based on torrefied wood pellets (external torrefaction). The heat integration as well as power consumption and production are shown in the flow sheet.

Table 5
Stream compositions for the methanol plant shown in Fig. 9 (external torrefaction).

	Gasifier exit	WGS outlet	AGR outlet	Reactor inlet	Reactor outlet	Purge gas	To distillation	Recycle CO_2
Stream number	7	11	16	18	19	26	29[a]	31
Mass flow (kg/s)	187.2	221.4	77.2	173.6	173.6	3.0	74.2	2.9
Mole flow (kmole/s)	8.76	10.66	6.95	13.64	9.21	0.21	2.31	0.08
Mole frac (%)								
H_2	26.1	42.9	65.8	61.1	42.2	56.3	0.27	8.2
CO	51.9	21.2	32.6	30.6	21.3	28.4	0.24	7.4
CO_2	8.9	28.7	1.2	3.0	4.3	4.9	2.5	75.9
H_2O	12.9	7.0	–	0.00	0.16	0.00	0.64	0.00
CH_4	0.03	0.02	0.05	0.56	0.84	1.1	0.05	1.5
H_2S	0.02	0.02	–	–	–	–	–	–
N_2	0.14	0.11	0.19	2.9	4.4	5.8	0.05	1.5
Ar	0.07	0.06	0.11	1.5	2.2	2.9	0.09	2.6
CH_3OH	–	–	0.03	0.35	24.6	0.68	96.1	2.9

[a] Liquid.

Table 6
Main results from the modeling and analysis of the two biorefineries.

	Integrated torrefaction		External torrefaction	
Biomass input [MWth]	500		2906	
Torrefied biomass input [MWth]	–		2312	
Electricity consumption:	[MWe]	[% of biomass input]	[MWe]	[% of torrefied biomass input]
Air separation unit	11	2	48	2
Milling	1	~0	7	~0
Compressors	11	2	49	2
Acid gas removal (AGR)	5	1	24	1
Total electricity consumption	28	6	127	5
Electricity production:				
Steam cycle[a]	14	3	243	11
Gas turbine[b]	14	3	18	1
Total electricity production	28	6	261	11
Net electricity production	0	0	134	6
Energy efficiencies [%]:				
Biomass to syngas (Overall cold gas efficiency)	86		63	
Torrefied biomass to syngas	–		80	
Syngas to methanol	73		77	
Biomass to methanol	63		49	
Biomass to net electricity	0		5	
Biomass to methanol + net electricity (total efficiency)	63		53	
Torrefied biomass to methanol	–		61	
Torrefied biomass to methanol + net electricity	–		67	

[a] The thermal efficiency of the steam cycles range from 36% for a simple cycle (600 °C and 43 bar) to 42% for a cycle with reheat (600 °C and 180 bar) – steam extraction is not included in these values.

[b] The thermal efficiency of the gas turbine is 37.9%.

no net electricity production and more difficult transport, storage and handling of the biomass feedstock (Table 7). An economic analysis would be necessary to determine which of the two biorefineries would be able to produce methanol at the lowest cost.

Two alternative designs for the biorefinery with integrated torrefaction are discussed below:

1) Increased methanol efficiency, 2) Atmospheric torrefaction and gasification.

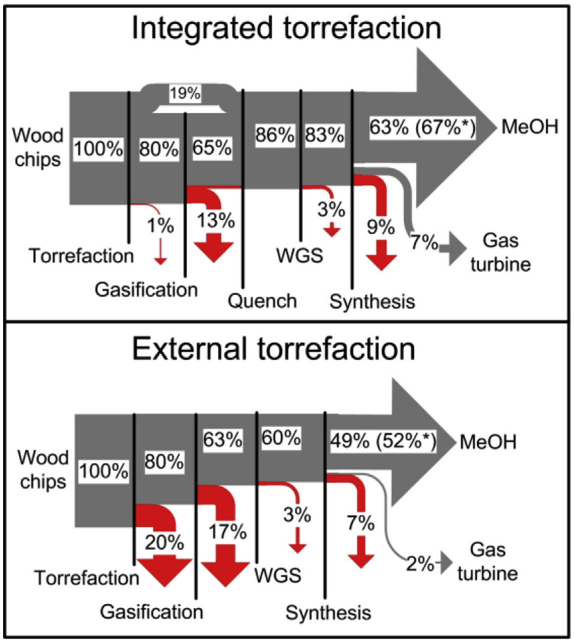


Fig. 10. Chemical energy flows (LHV-dry) and conversion heat losses (red) in the two biorefineries. Note: the torrefaction process occurs outside the plant for "external torrefaction". * The value given in parentheses is the energy value of methanol in the gas state (LHV). If this value is used, the width of the arrows before and after synthesis would be equal. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

1) Increased methanol efficiency: The methanol efficiency could be increased by increasing the recycle ratio in the synthesis loop from 88% to 97%, which matches the recycle ratio used for external torrefaction. The syngas to methanol efficiency would thus be raised from 73% to 77%, which also matches the syngas to methanol efficiency for external torrefaction (Table 6). This increase would increase the biomass to methanol efficiency

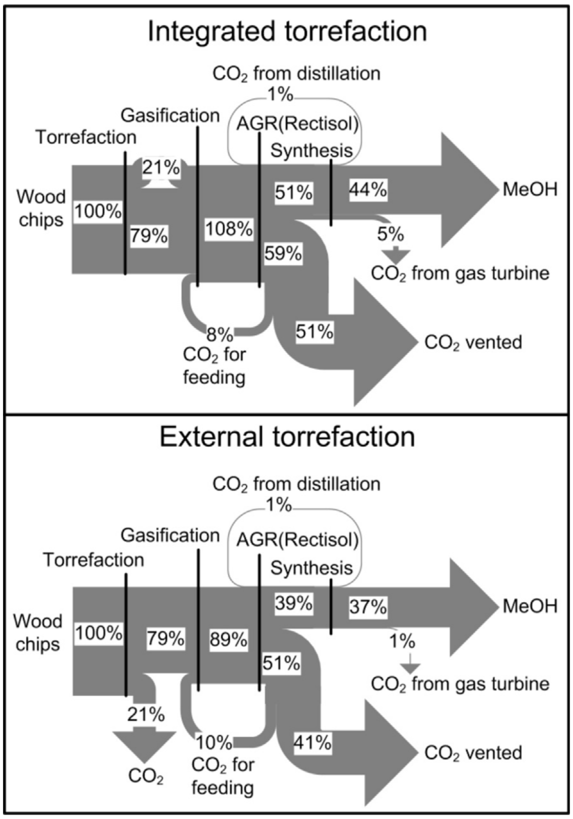


Fig. 11. Carbon flows in the two biorefineries.

Table 7
Pros and cons for the two investigated biorefineries.

	Pros	Cons
Integrated torrefaction	1. Increased energy efficiency	1. More difficult transport, storage and handling of the biomass feedstock (wood chips) 2. Smaller plant (economy of scale) 3. No net electricity production 4. More complex plant design[a]
External torrefaction	1. Easy transport, storage and handling of the biomass feedstock (torrefied wood pellets) 2. Large plant (economy of scale) 3. Net electricity production	1. Reduced energy efficiency

[a] "More complex plant design" refers to: 1. the integrated torrefaction, e.g., pressurized torrefaction and/or pressurization of hot volatiles from torrefaction, 2. More complex heat integration in the plant (compare Fig. 8 with Fig. 9).

from 63% to 66%, but electricity would have to be imported to the plant as a consequence.

2) Atmospheric torrefaction and gasification: If atmospheric torrefaction and gasification are used as proposed in Ref. [4], the pressurization of solid wood prior to torrefaction and/or hot volatiles from torrefaction would be avoided. This approach would reduce the complexity of the integrated torrefaction. The most important consequence would be a 6-fold increase in the electricity consumption of the syngas compressor (to 5% of the biomass energy input). If the electricity consumption is to be met on-site, the syngas consumption of the gas turbine would increase. This increase would reduce the total efficiency from 63% to 57%. This efficiency is only slightly higher than the total efficiency of external torrefaction (53%). Because electricity typically is valued higher than fuel, the difference in the total efficiency would be insignificant.

For external torrefaction, integrating waste heat from another source (e.g., industrial processing plant) in the production of torrefied wood pellets to dry the biomass would greatly increase the energy efficiency of torrefaction from 80% (wood to solid product efficiency for torrefaction at 300 °C) up to 96% (wood to solid product efficiency for torrefaction at 250 °C).

6. Conclusion

Integrated and external torrefaction were analyzed and compared via thermodynamic modeling. The first part of the analysis showed that the biomass to syngas efficiency can be increased from 63% to 86% (LHV-dry) when switching from external torrefaction to integrated torrefaction at 300 °C. It also showed that integrated torrefaction at 250 °C and gasification without torrefaction yield biomass to syngas efficiencies of 81% and 76%, respectively. The increase in biomass to syngas efficiency when increasing the torrefaction temperature from 250 °C to 300 °C for integrated torrefaction was mainly due to 1) volatiles avoiding the gasifier and 2) the chemical quench that occurs when the volatiles are mixed with the hot gas from the gasifier. The increase in cold gas efficiency of the gasifier only contributed slightly to the increase in the overall biomass to syngas efficiency. The second part of the analysis showed that the total energy efficiency of a biorefinery could be increased from 53% to 63% (biomass to methanol + net electricity) when switching from external torrefaction to integrated torrefaction at 300 °C. This increase in energy efficiency incurs the following costs: 1) More difficult transport, storage and handling of the biomass feedstock (wood chips vs. torrefied wood pellets), 2) reduced plant size, 3) no net electricity production and 4) a more complex plant design. A detailed economic analysis is needed in order to decide which of the two biorefineries that would be most cost-effective.

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